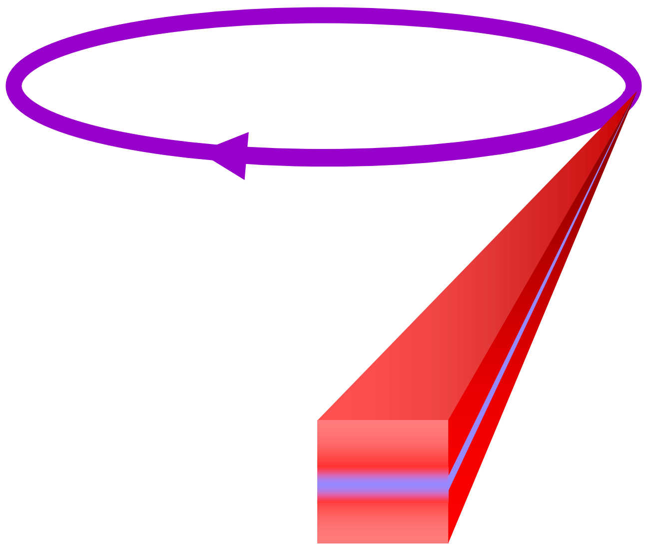
Infrared Spectroscopy of Solids at the NSLS

J. Hwang, H. Zhang, H. Tashiro, J. LaVeigne,
D.H. Reitze, C.S. Stanton, and D.B. Tanner
University of Florida
R.P.S.M. Lobo
ESPCI
Laszlo Mihaly
Stony Brook
Jiufeng Tu
City College of New York
G.L. Carr
NSLS

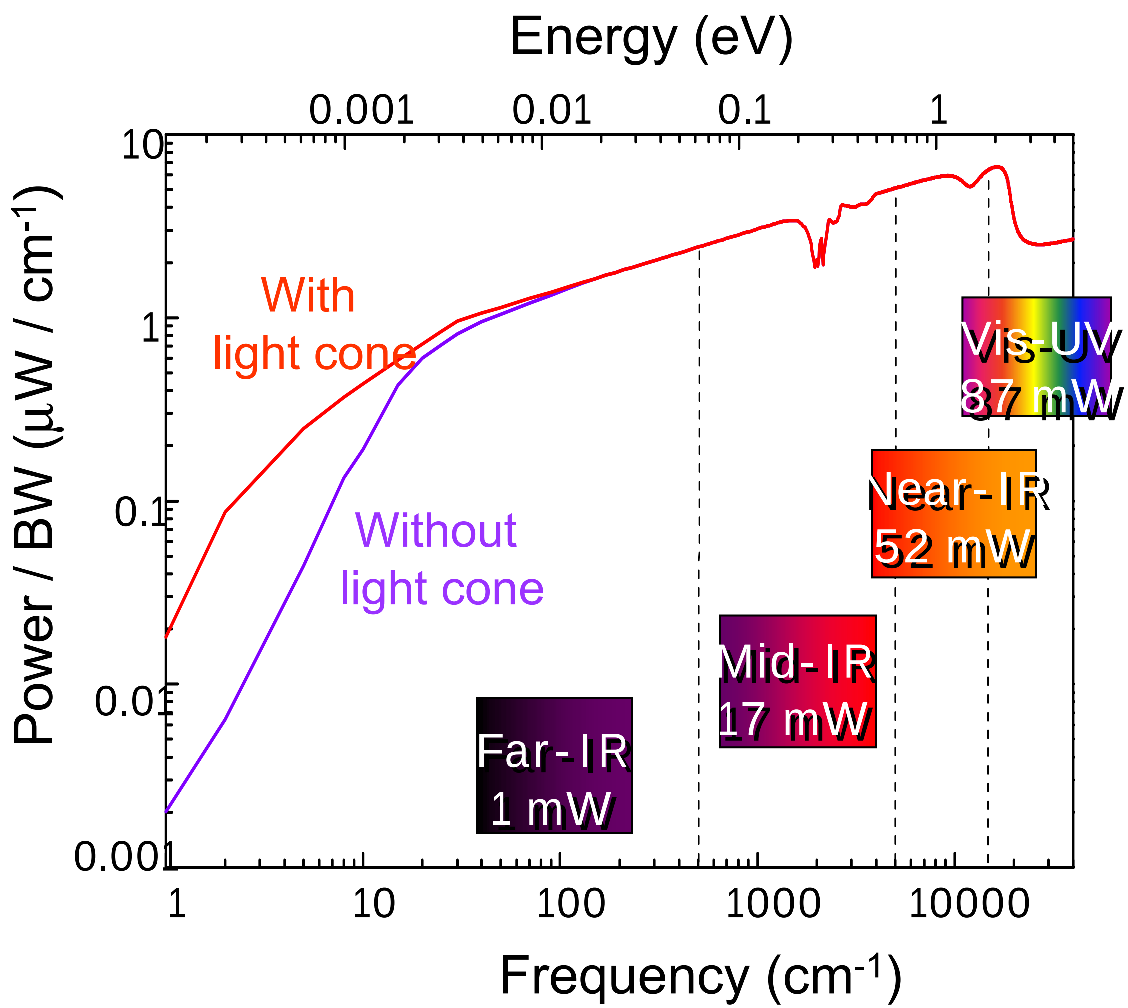
Outline

- Measurements in the very far IR
  - mm waves, terahertz
  - Physics opportunities
  - Needed technology
- Nonlinear far-IR spectroscopy
  - Time-resolved (pump-probe)
    - Metallic superconductors
    - Magnetic semiconductors
  - High *E*-field-strength opportunities

Infrared Synchrotron Radiation



- “White” source
- High Brightness
- Long wavelength flux (the far-infrared)
- Pulsed (100s of ps)



Physics in the far-IR

Important physics ("All of solid-state physics…" A.J. Heeger

- Superconducting gaps
- Antiferromagnetic and ferromagnetic resonance
- Collective modes
- Free-carrier dynamics

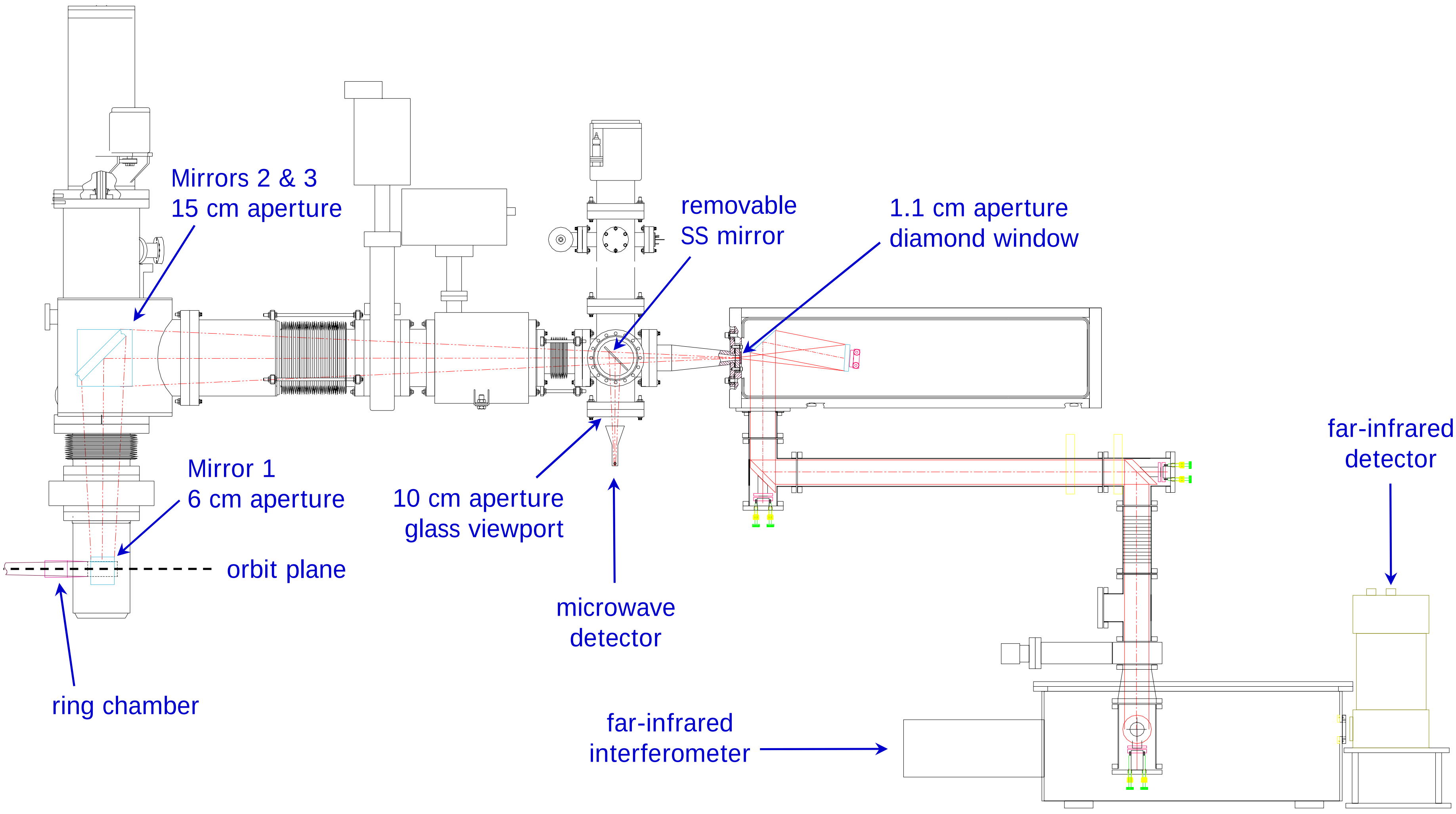
Materials

- Quasiparticles in HTSC
- Gaps in LTSC
- Heavy Fermions
- CDW ($TaSe_2$)
- Organics ($TMTSF_2$-X, $BEDT\text{-}TTF_2$-X) (CDW, SDW, SC)
- Dilute magnetic semiconductors

Spectroscopically difficult

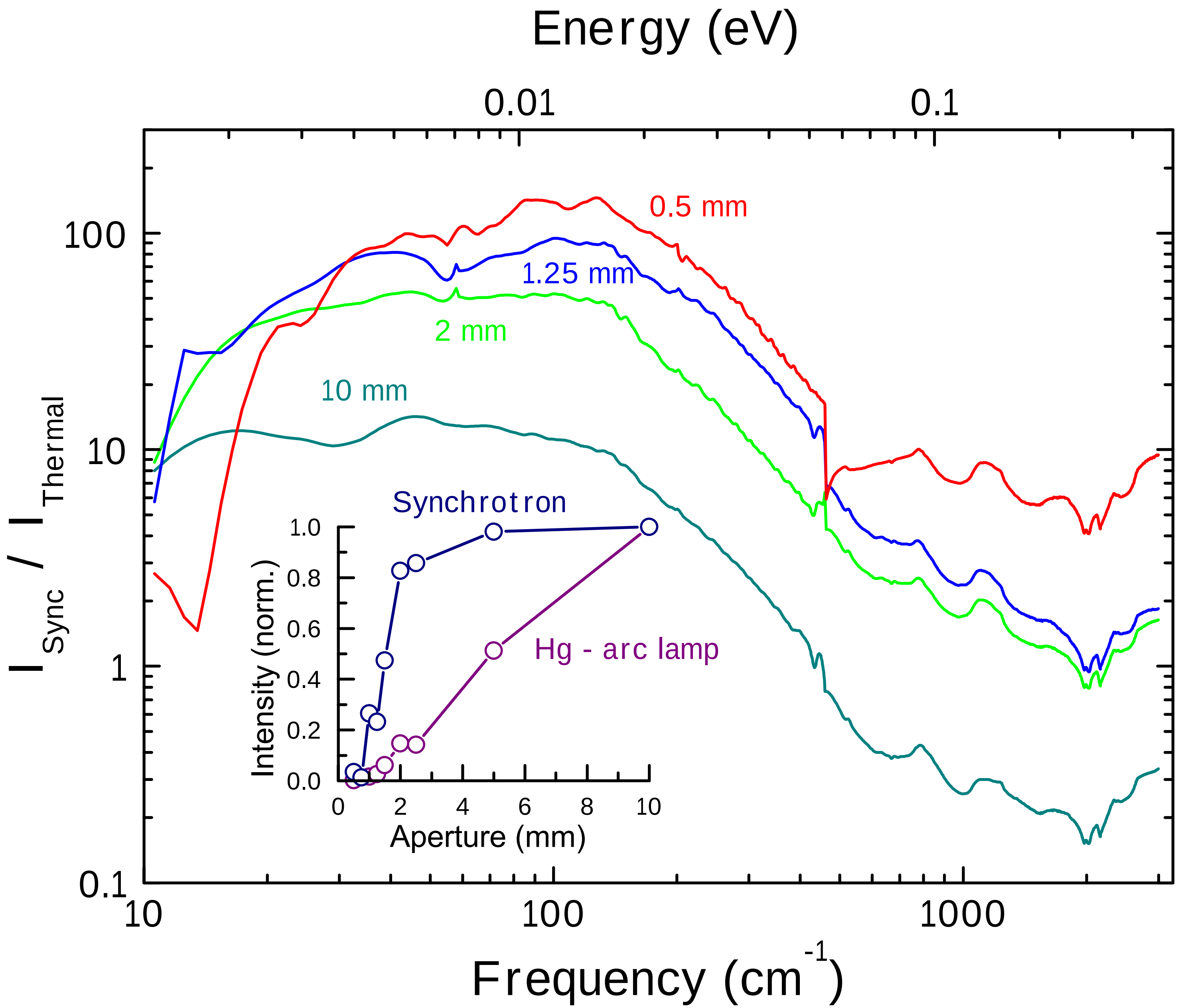
- Weak thermal sources for FTIR
- High frequencies for microwave sources
- THz generators not mature, limited in bandwidth
- Synchrotron sources, therefore, have advantages.

U12IR Beamline on Dipole Bend



Typtical Beamlines

Synchrotron & Thermal Sources



Signal to noise comparison

Synchrotron radiation allows

low noise far-IR measurements.

IRSR is diffraction limited.

but…

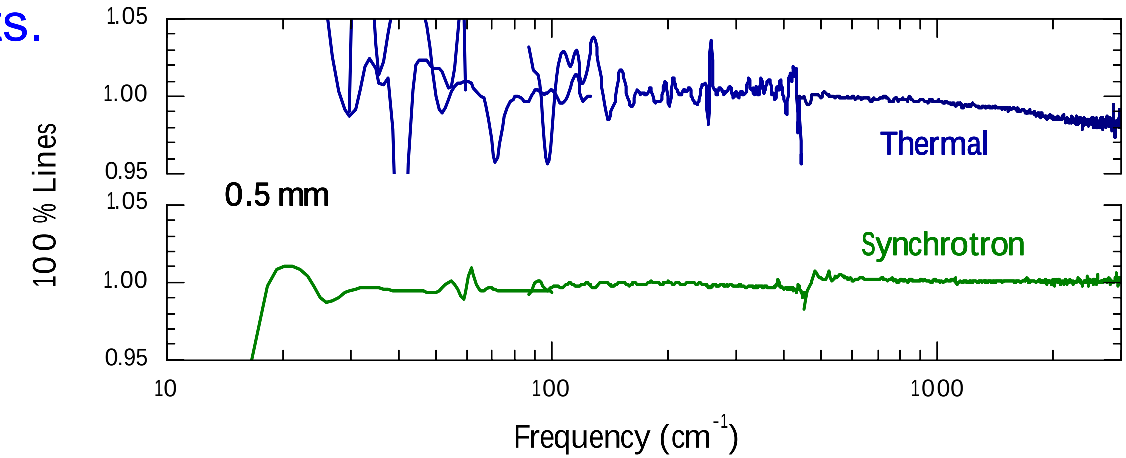
Beam noise has to be

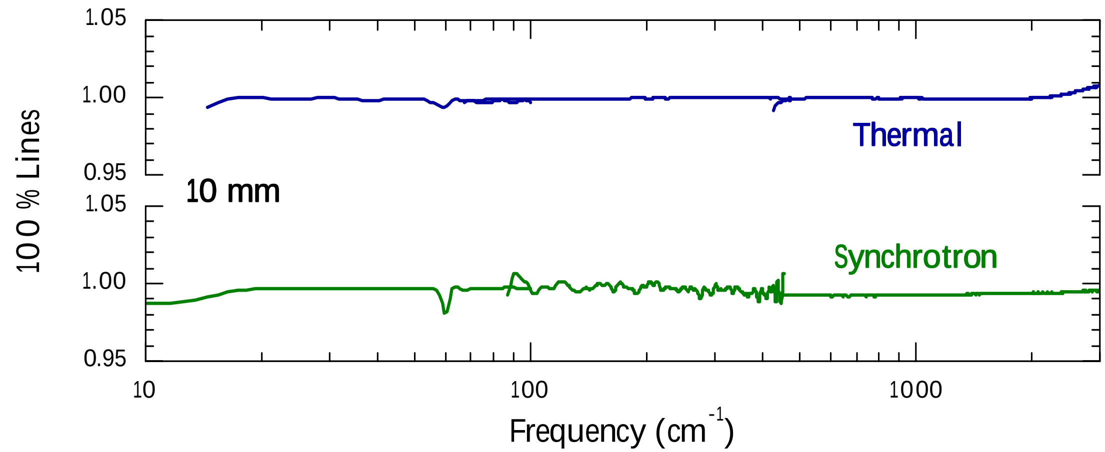
taken into account for larger

samples.

(Note also the effect of aperture

diameter at low frequencies.)

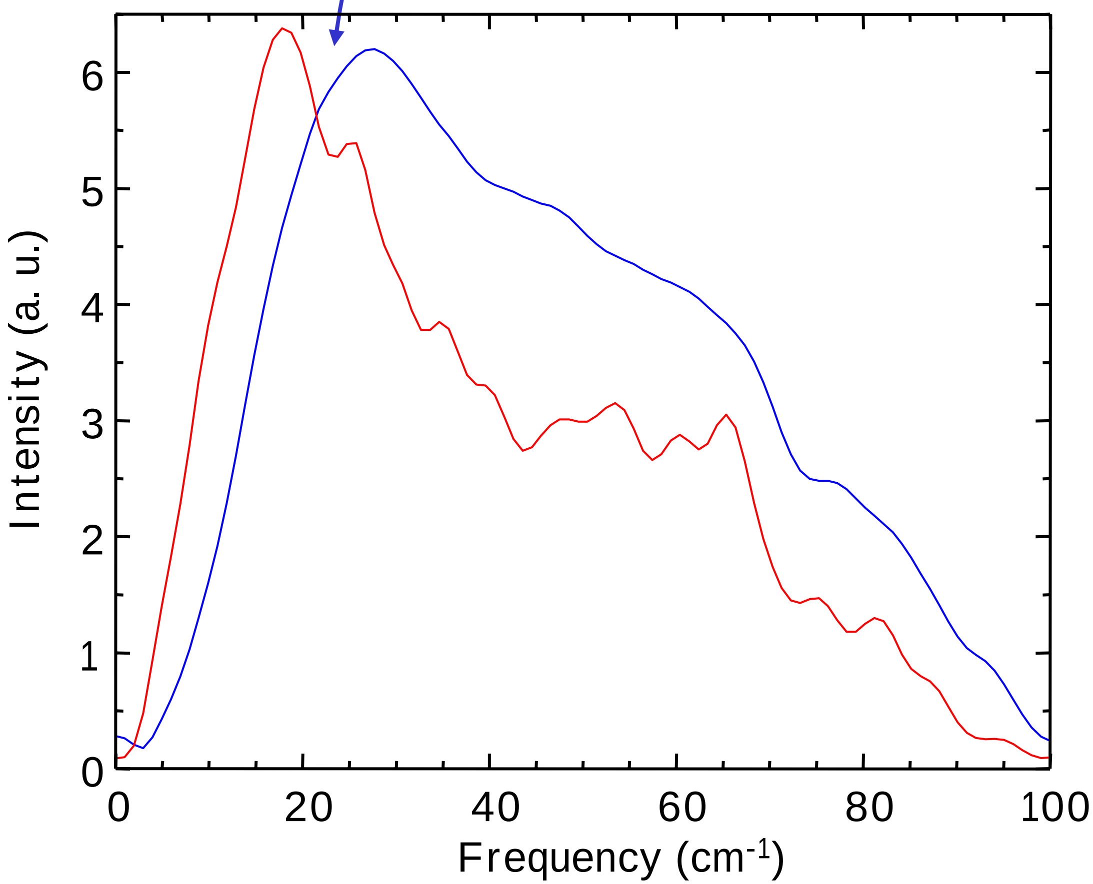




Synchrotron vs. UA-3 Hg arc

Comparison uses ½” aperture, lamellar grating interferometer

Crossover between Hg arc lamp and synchrotron radiation at ~ 20 cm-1



Notions about design requirements

1. Efficient transfer of light from ring to detector
2. Spectroscopic approach
3. SNR of detector
4. Sample temperature range
5. Technical noise from ring
6. Interference, standing waves

Notions about design requirements 2

1. Efficient transfer of light from ring to detector
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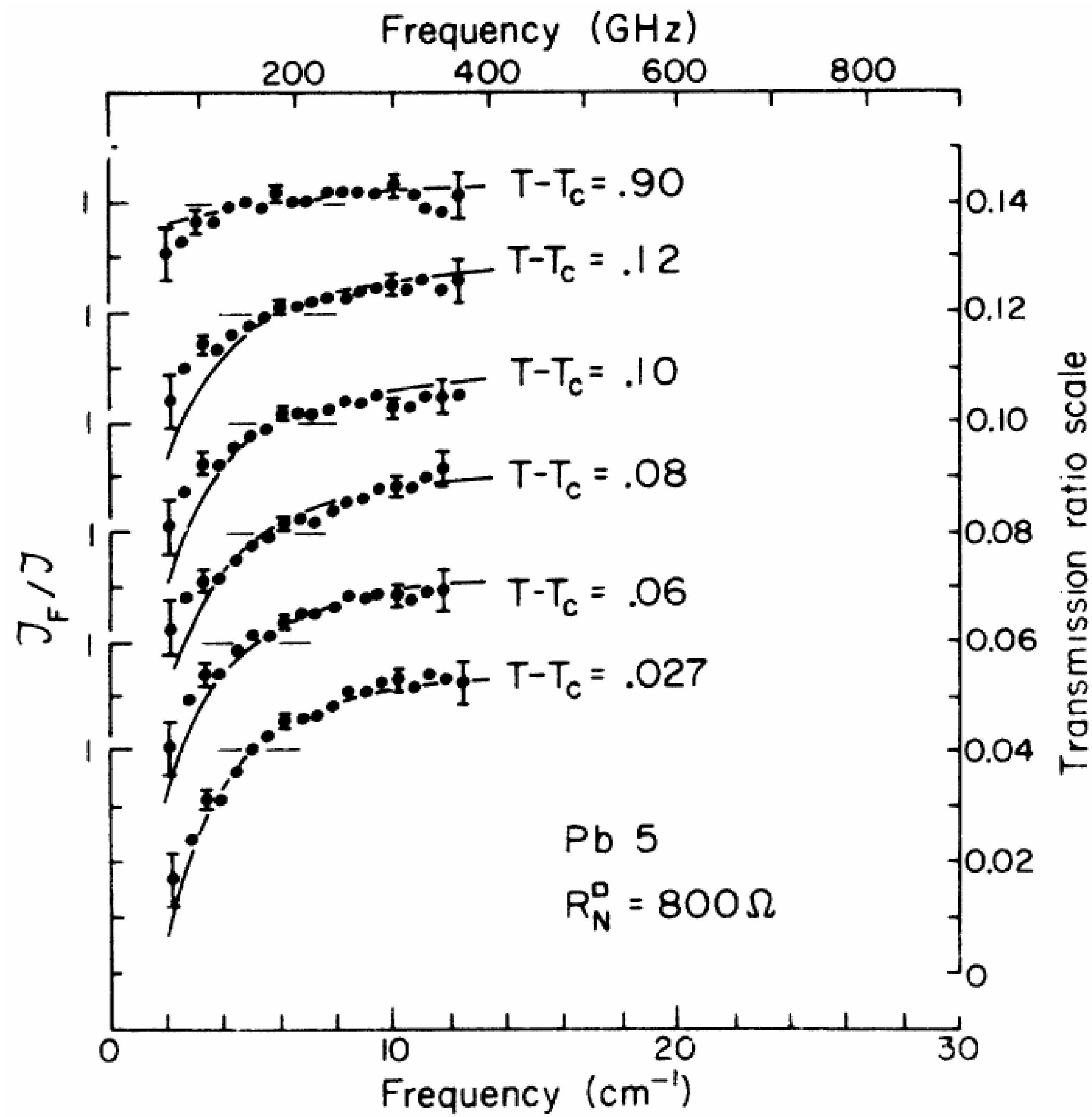
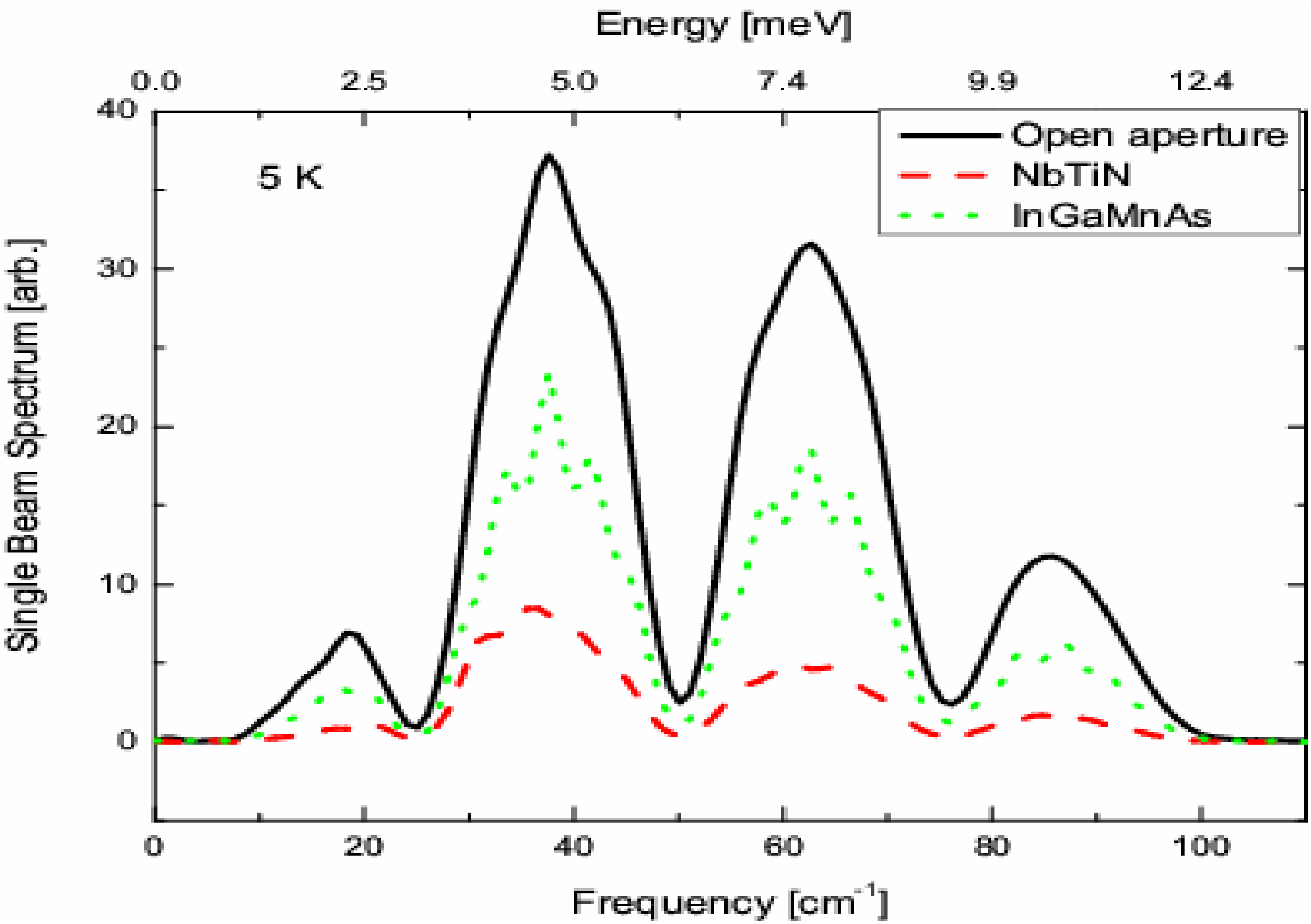
6. Interference, standing waves

- With no sample, transfer 75% of light
- FTS, then R-> KK or R+T or ellipsometery. Magnet, pressure.
- Best detector available, use 0.3 K
- Get $T < \nu$
- Active bunch steering, active beam steering
- Dump reflected beams, aim for low VSWR.

1971 State of the art



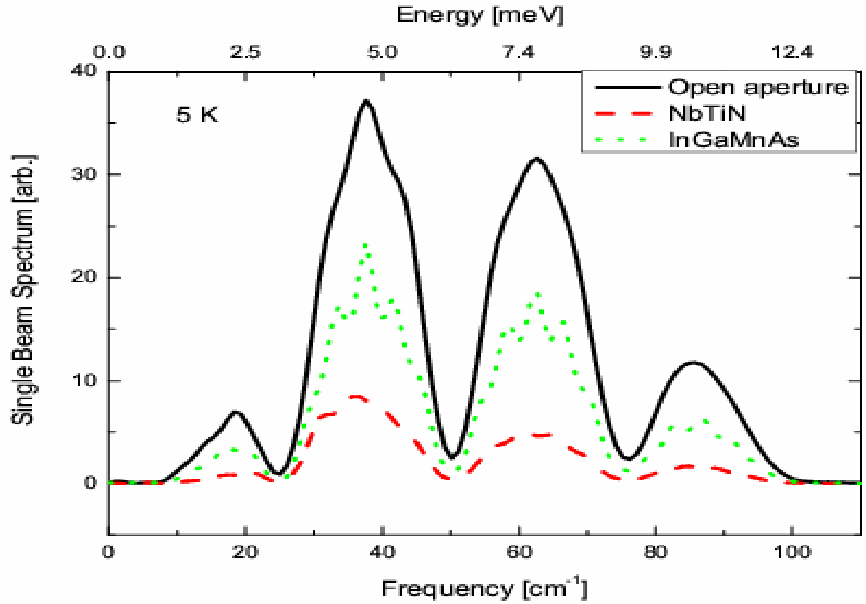
High pass filter in detector?



High pass filter in detector? 2

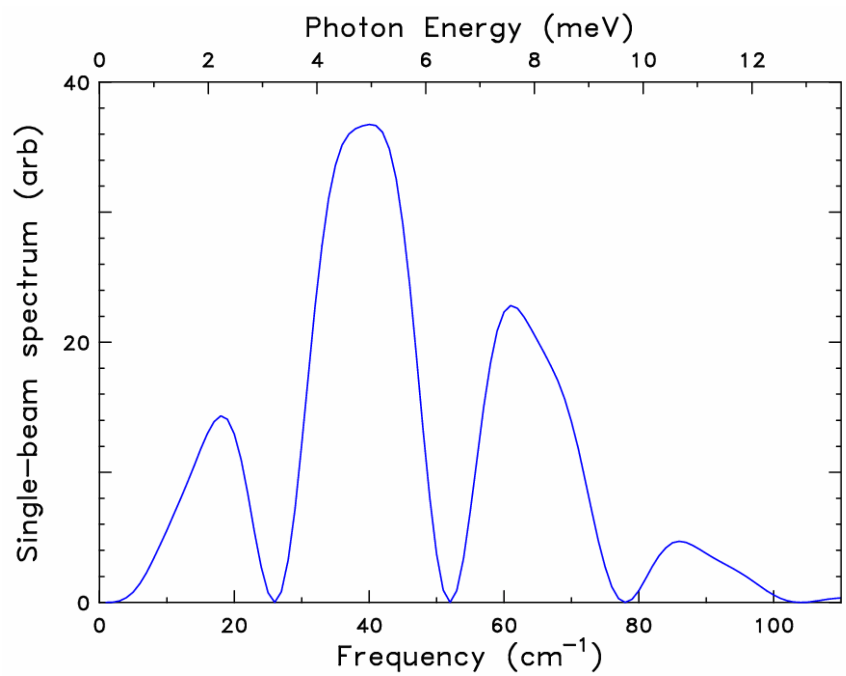
Simulated spectrum:

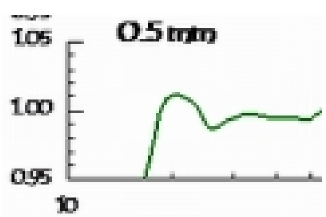
- Mylar beamsplitter, with fringes
- Efficiency $\varepsilon = 4RT$
- $S(\nu) \sim \nu^2$ (Rayleigh-Jeans)
- Long pass filter with 100 cm^{-1} cutoff
- $\alpha = C\nu^2$



Simulation has *much* more low-frequency energy than what is observed.

(4x at 10 cm^{-1}, 12x at 8 cm^{-1})



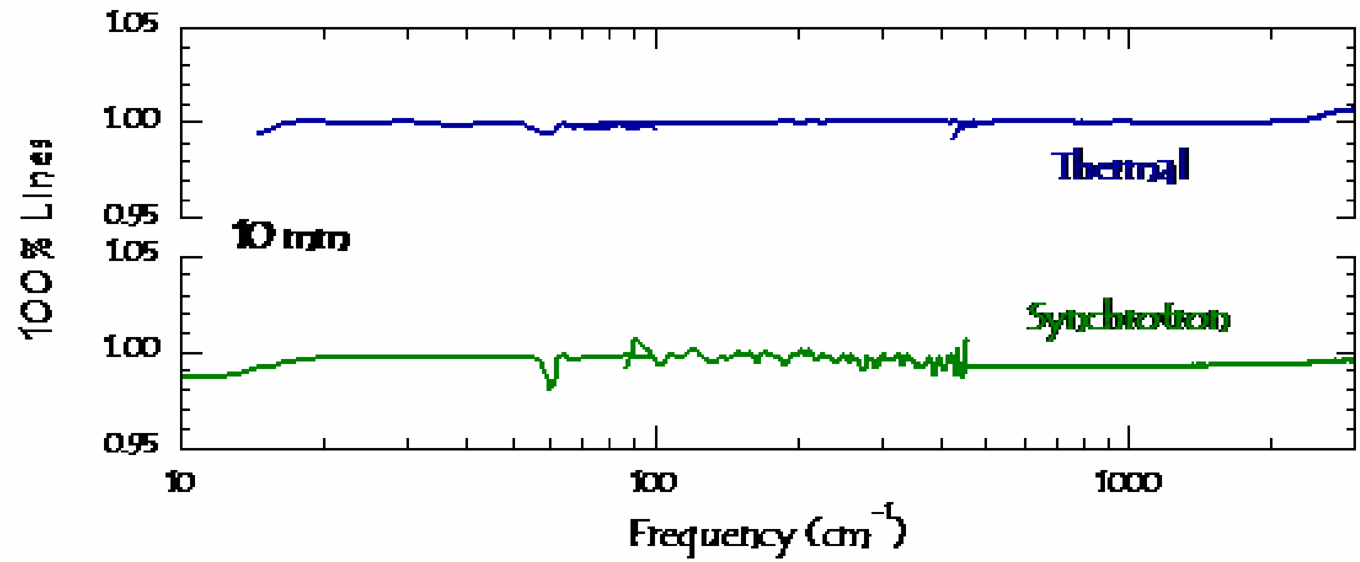


Need for lower temperatures

- Take $h\nu = kT$. (1 cm^{-1} ⇔ 1.44 K)
- Assume feature of interest is a mean-field gap at 5 cm^{-1}.
- Then T_c = 2 K, taking $2\Delta = 3.5T_c$
- Thus, 200 mK is $T_c/10$.

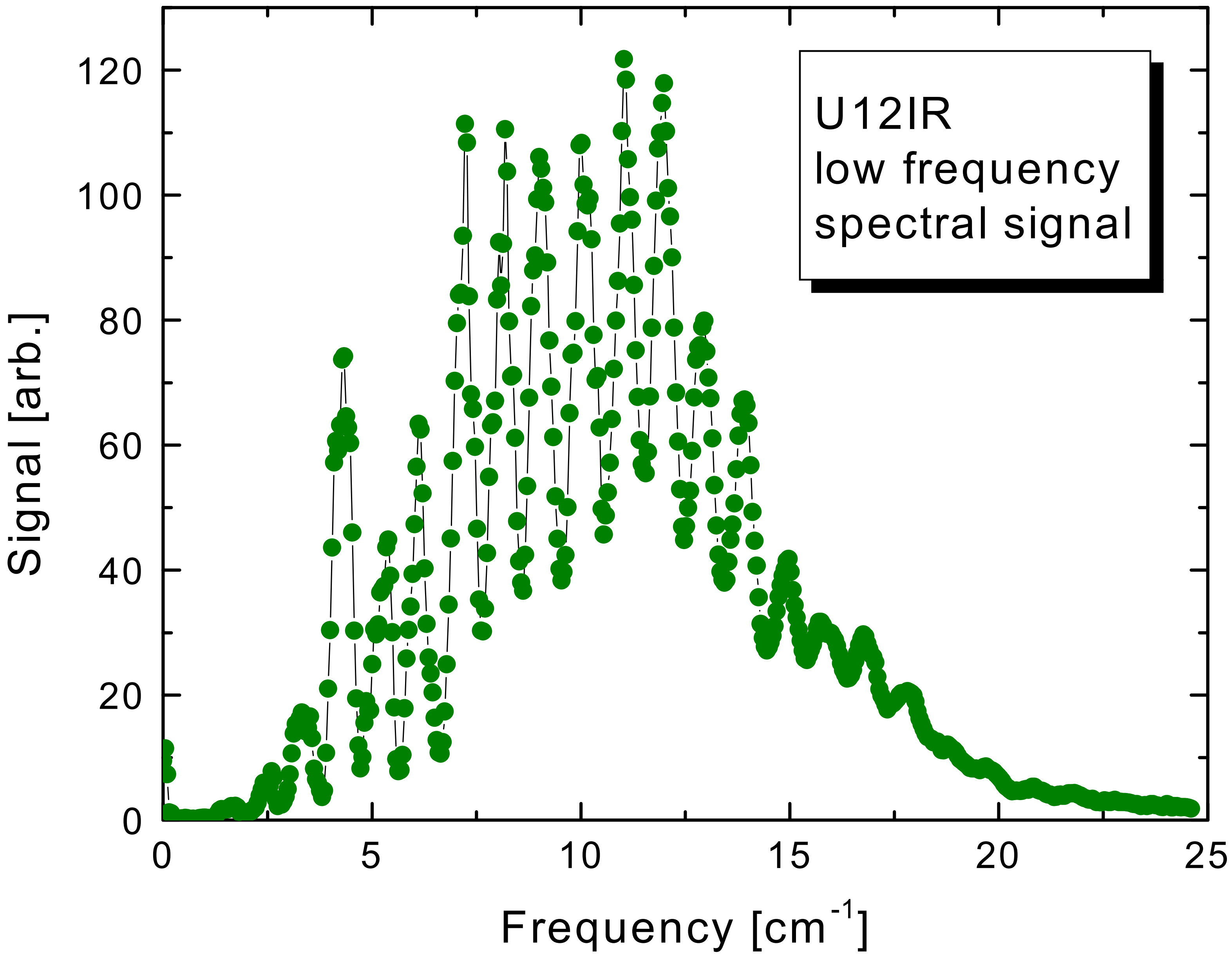
- Options (low-cost to high-cost):
  - Attach sample at bottom of ^3He cryostat for 0.3K detector (e.g., Reedyk, Basov, others)
  - Build ^3He fridge, one shot or recirculating, in Dewar with optical windows.
  - Dil fridge. Cryogen free, easy operation for ~200k.

Beam Jitter



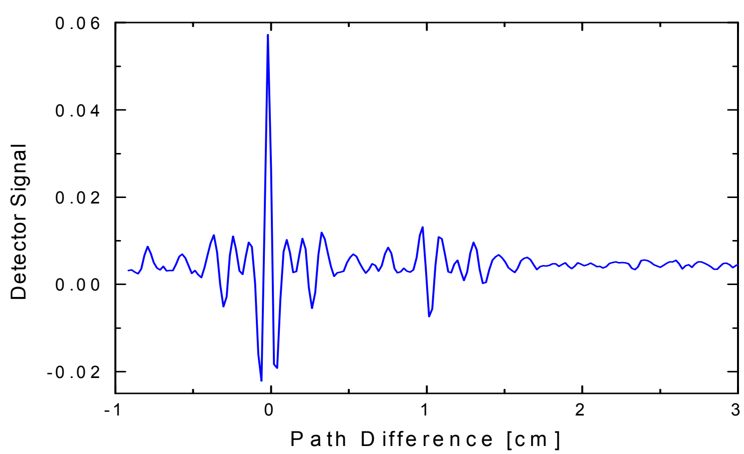
- Need to reduce technical noise from SR.
- Seems to be due to beam jitter
- Use quad photodiodes at conjugate image planes, feed back to steering mirrors (e.g., LIGO)
- Could use beam divider, measure incident intensity, divide
- Throughput requirements would not be met

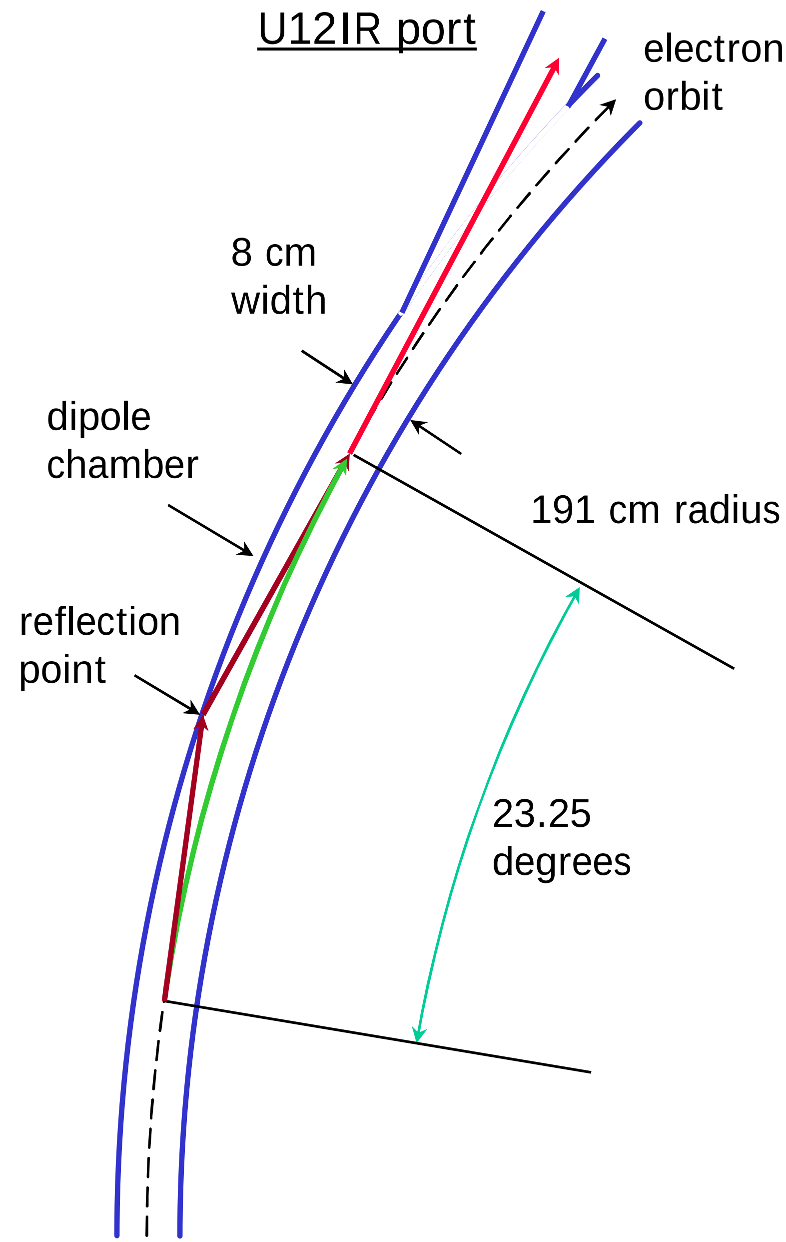
U12IR fringe pattern



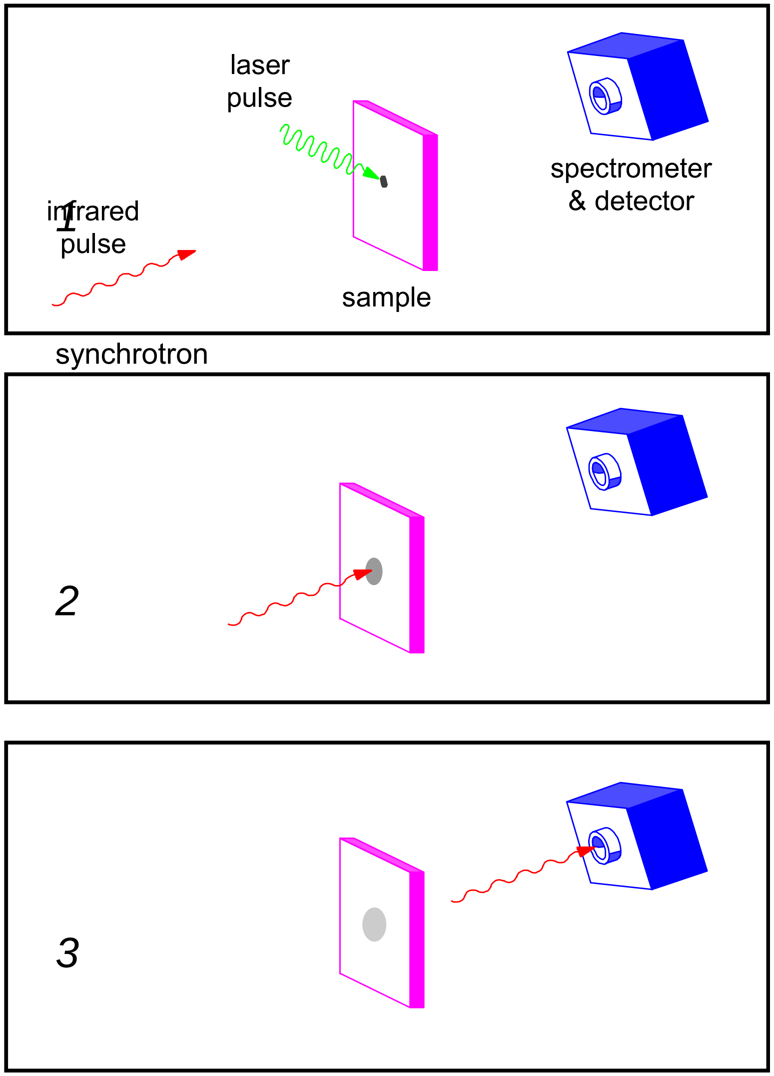
"Echo" produced by dipole chamber

- Need to absorb wall reflections
- Light reflected from wall enters interferometer with well-defined phase wrt prompt beam.
- Affected by beam motion, chamber temperature!
- Gives second signature in interferogram.





Time-resolved IR



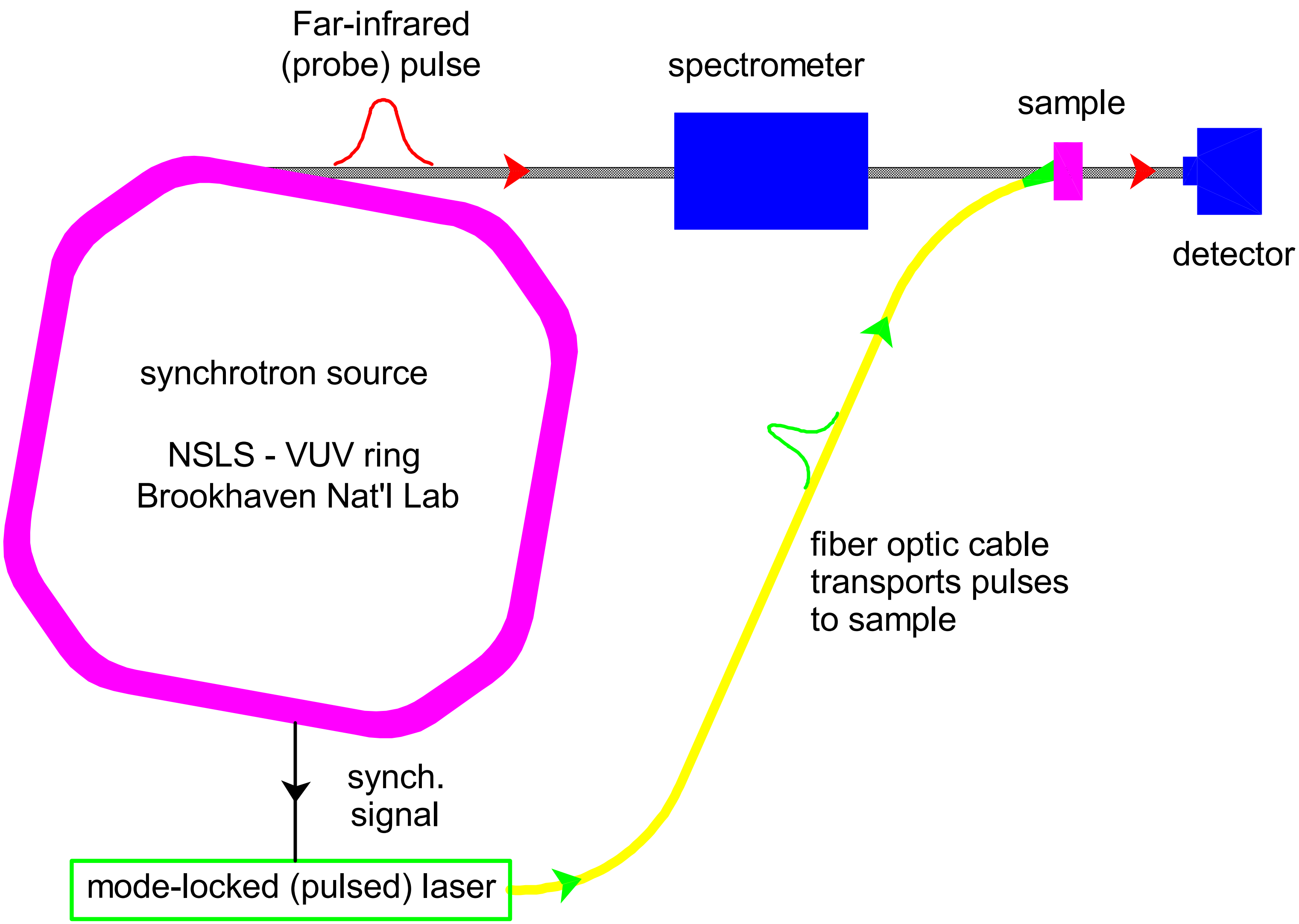
1. Laser pulse creates photoexcitations in sample, which subsequently evolve with time.

2. After time Δt, broadband (continuum) IR pulse arrives and is partially absorbed (or reflected) by excitations.

3. IR pulse analyzed with a spectrometer, extracting details of excitations at a time Δt after their creation.

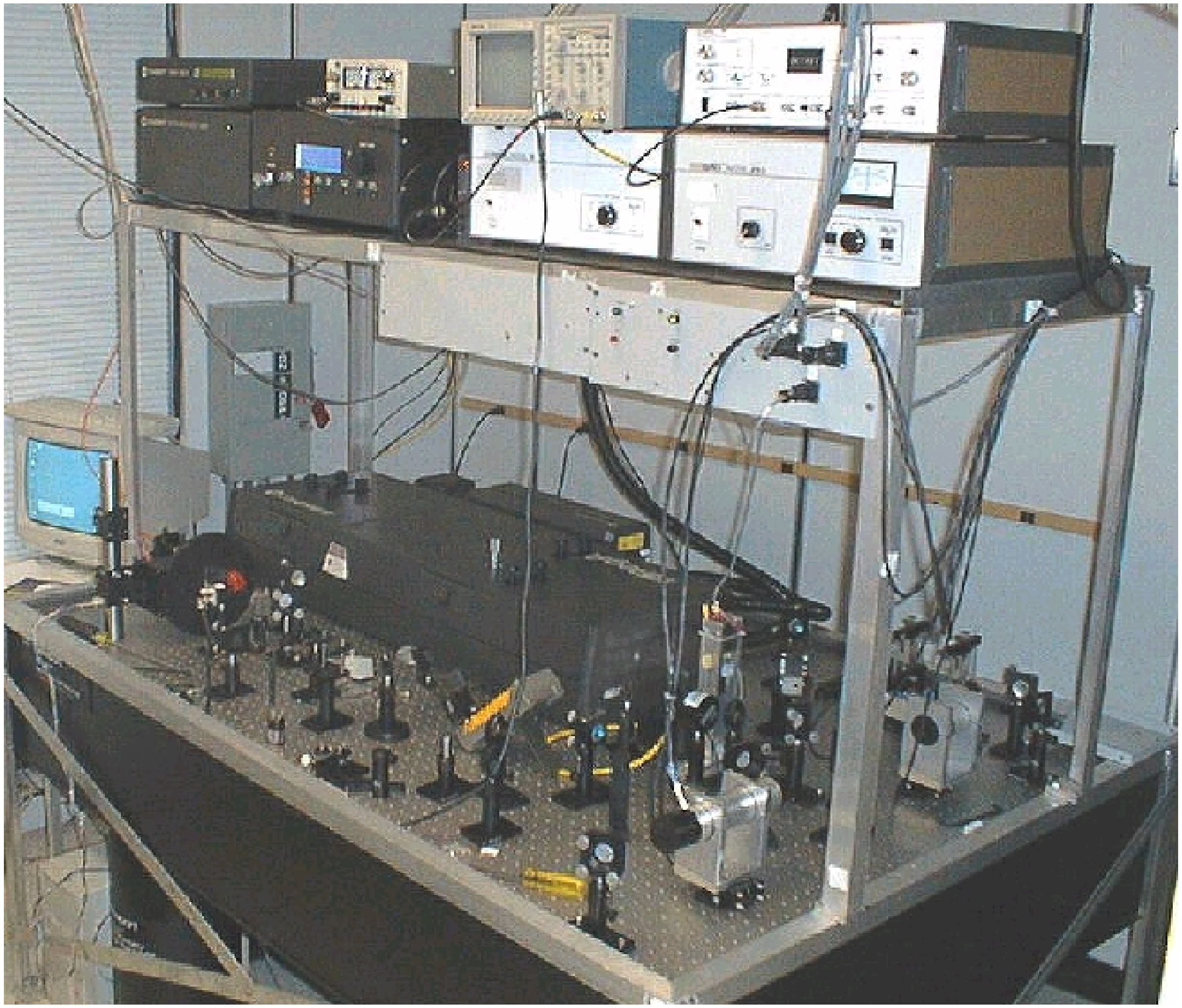
- Cycle repeats at high (50 MHz) repetition rate.
- Photoexcitation evolution determined by measuring at a variety of Δt's.
- Employs "conventional" spectroscopy using high-sensitivity (slow-response) detectors.

Pump-probe with Laser & Synchrotron Pulses



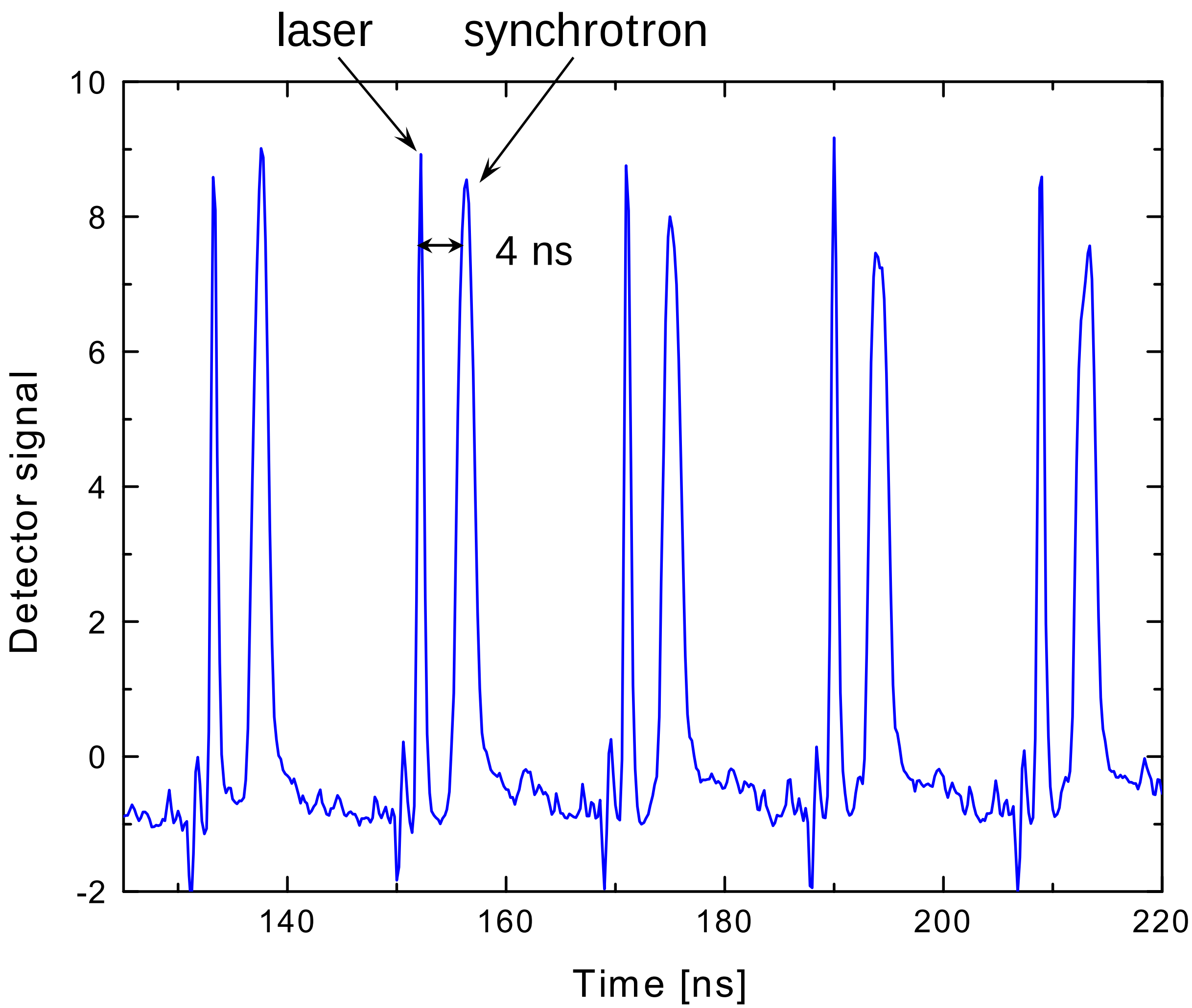
Synchronized Ti:sapphire laser

- Coherent mode-locked laser (MIRA 900p) pumped by Verdi green laser
- Tunable (700-1000 nm)
- Frequency doubling capable
- ~ 800 mW average power
- 2 ps pulses
- PRF synchronized to 2x NSLS 53 MHz RF system (VUV or X-ray)
- Pulse selection to match various synchrotron bunch patterns
- Optical fiber delivery to beamline(s).



Pulses at specimen location

- Synchronized laser & storage ring pulses.
- Measured at sample location.
- Ge APD (near IR detector, ~1 ns response).
- Useful for locating "zero" delay point. Shown with 4 ns delay.
- Synchrotron pulse length ~300 ps (compressed mode)



- NSLS II: bunch length is 10 ps (ordinary mode)

Photoexcitation / Relaxation in Superconductors

high energy ($E \sim E_F$) electronic excitations (quasiparticles)

~ femtoseconds

low energy qp's & E_{Debye} phonons

$h\nu$

~ picoseconds

excess Δ qp's $\underset{1/\tau_B}{\overset{1/\tau_R}{\rightleftharpoons}}$ excess 2Δ phonons

Bottleneck => ns

phonon escape (at rate $1/\tau_\gamma$)

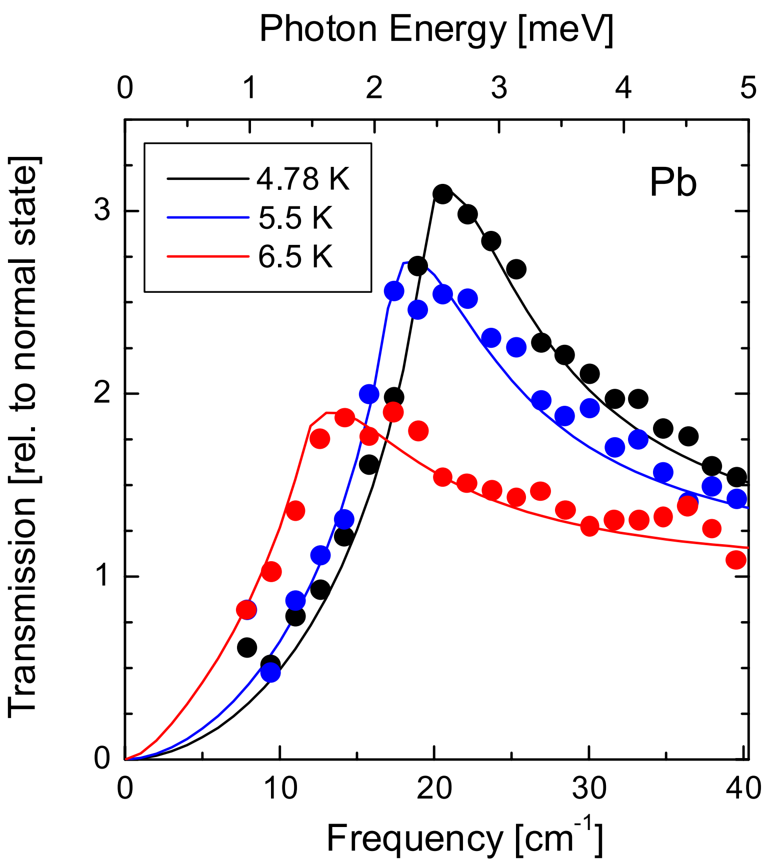
paired electrons & thermal quasiparticles

- Multi-step process, with a range of timescales.
- Coupled system of excess quasiparticles and phonons, trapping and effective lifetime.
  - Rothwarf & Taylor

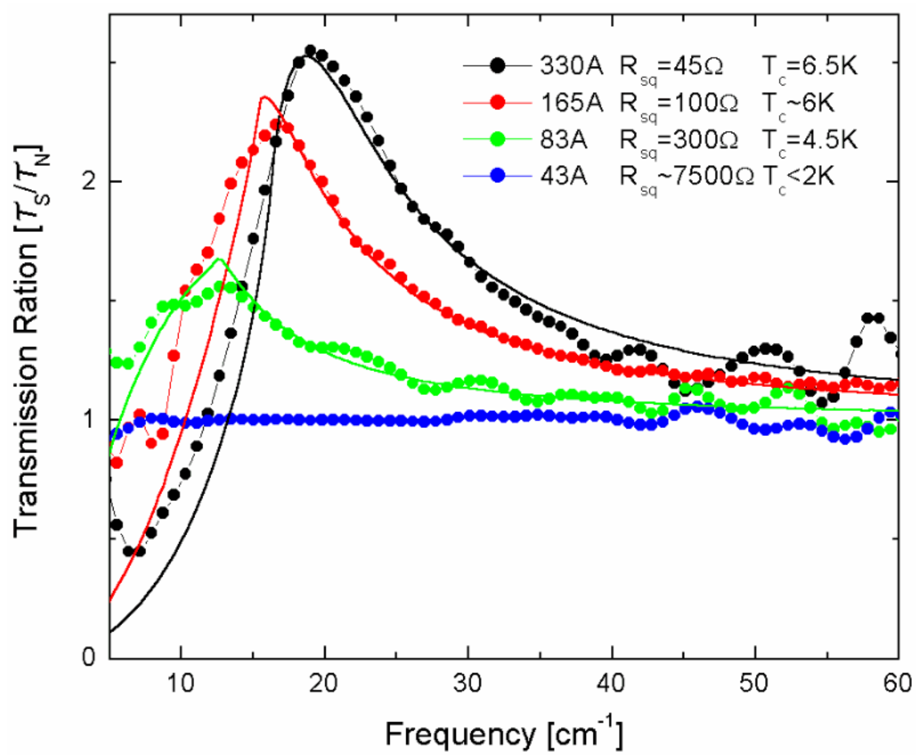
Far-infrared transmittance

Peak in T_s / T_n is a measure of the superconducting gap

Pb



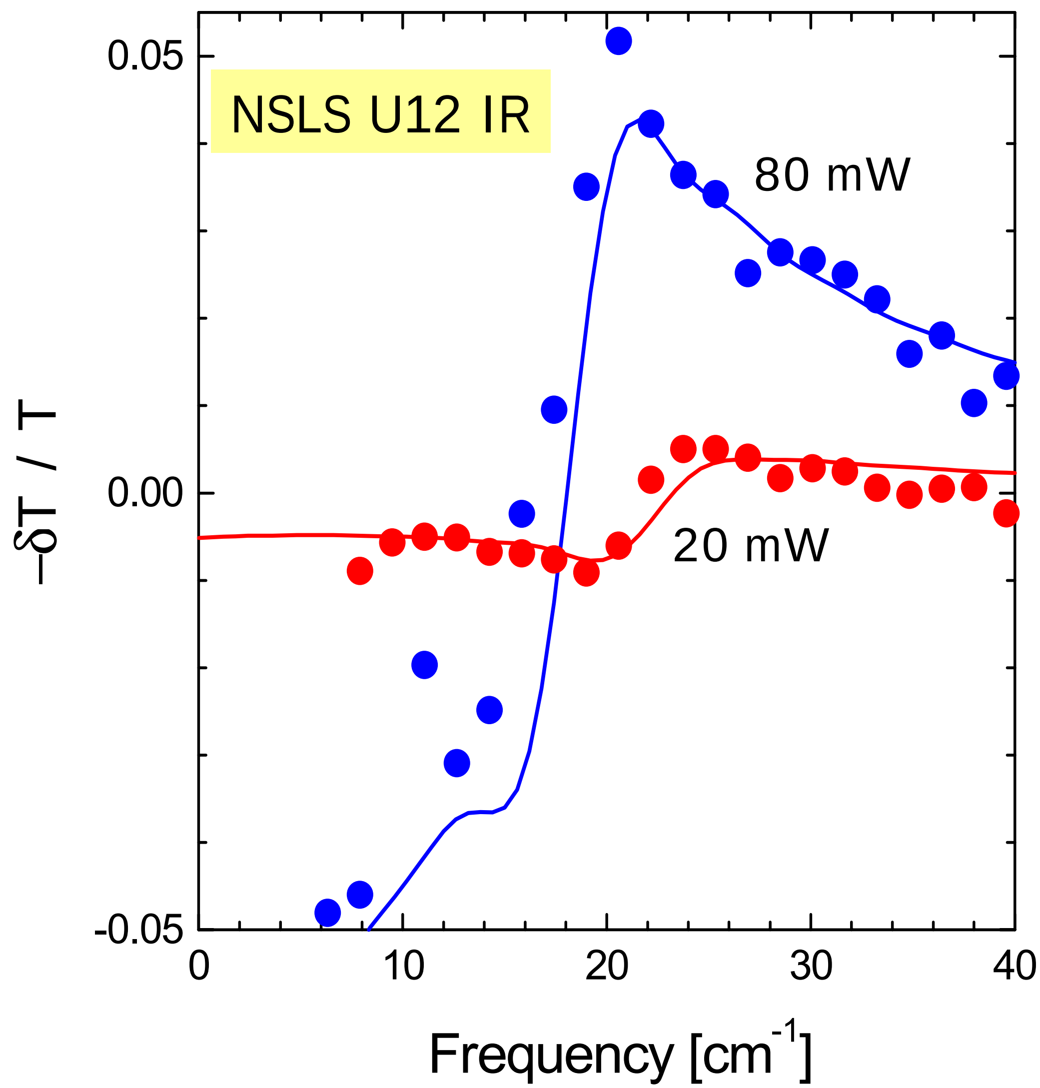
MoGe (at 2 K)



Energy gap shift from pair-breaking

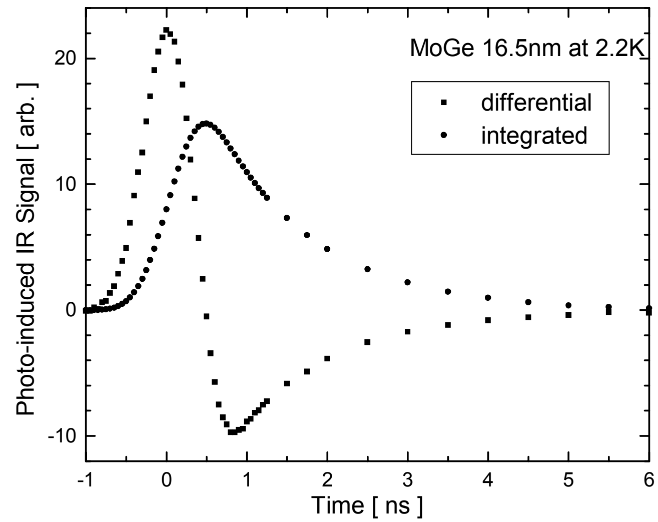
- Superconducting Pb film on sapphire.
- Experiment (solid circles) & theory fits (sold lines).
- Good agreement
- Gap shift (~ 0.7 cm^{-1} or 3%), sensed as change in far IR transmission.
- Density of excess q-p's <2%, comparable to or less than thermal population

=> weak to moderate perturbation



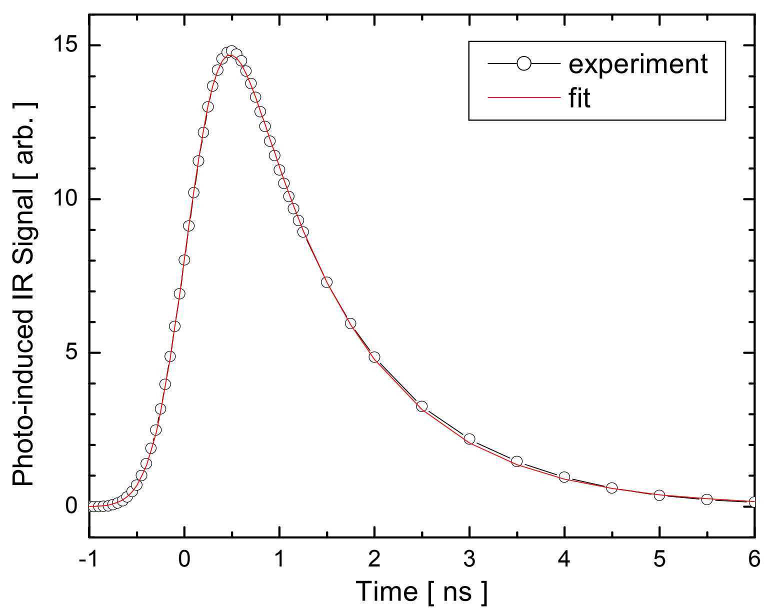
Time–dependent relaxation measurements

- Differential Technique:
  - pump-probe delay “dithered”.
  - differential transmittance signal (spectral average) for a range of delay time.
  - time-dependent relaxation of excess quasiparticles by integration.



- Relaxation Behavior:
  - convolution of simple exponential decay and Gaussian synchrotron pulse.
  - decay time ~ 1 ns.
  - time-resolution determined by synchrotron pulse width (~300 ps).

$$\Delta T = \frac{1}{2} A \exp\left(\frac{w^2}{4\tau^2} - \frac{t-t_0}{\tau}\right)\left(1 + erf\left(-\frac{w}{\tau} + \frac{t-t_0}{w}\right)\right)$$



Temperature dependence of relaxation time

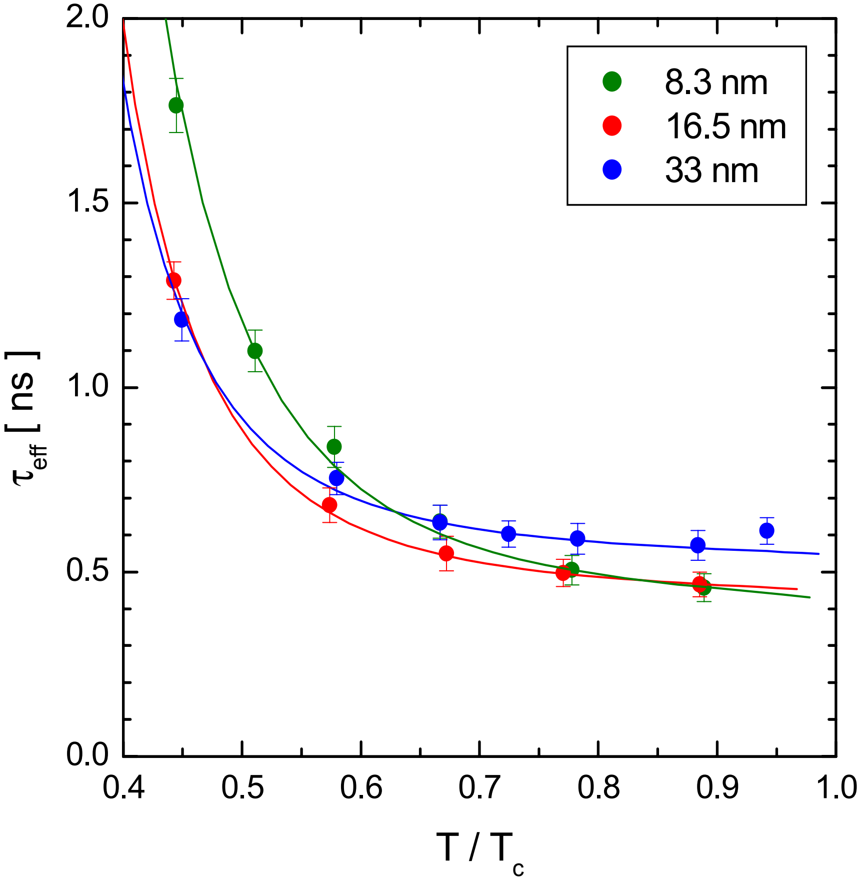
$$\tau_R(0) = (1+\lambda)\hbar / 2\pi b\,(kT_c)^3$$

$$\tau_B(0) = \hbar N_\Omega / 4\pi^2 N(0) \langle \alpha^2 \rangle_{av} \Delta_0$$

$$\tau_{eff} \approx \tau_\gamma + (1/2)\tau_R(1+\tau_\gamma/\tau_B)$$

- Relaxation times for MoGe films:

d [nm]	τ_γ [ps]	$\tau_R(0)$ [ps]	$\tau_B(0)$ [ps]	$\tau_R(0)/\tau_B(0)$
8.3	420	370	105	3.50
16.5	450	150	79	1.87
33	550	100	75	1.35

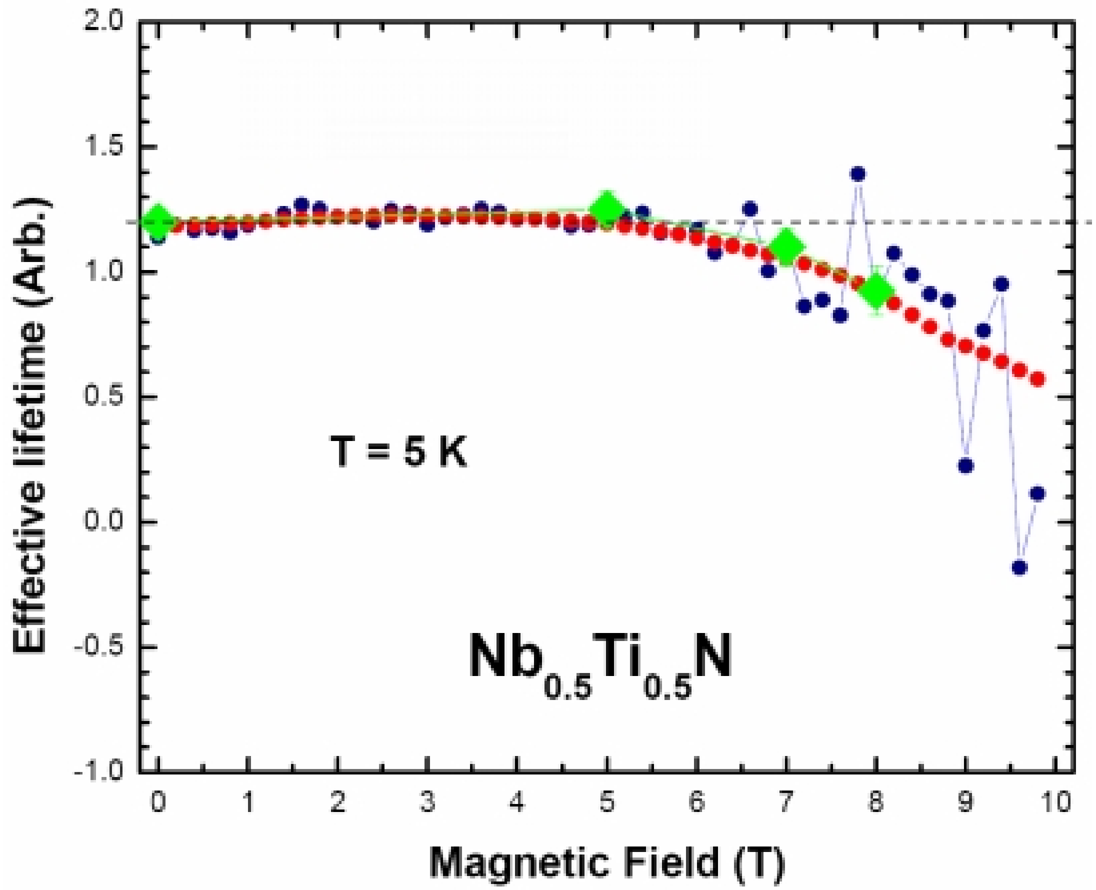


S. B. Kaplan et al.

Magnetic field

- Apply magnetic field
- Field puts vortex cores in the superconductor
- These give another relaxation channel
- Expect that the relaxation time will decrease with increasing magnetic field
- Follows the area density of vortices?

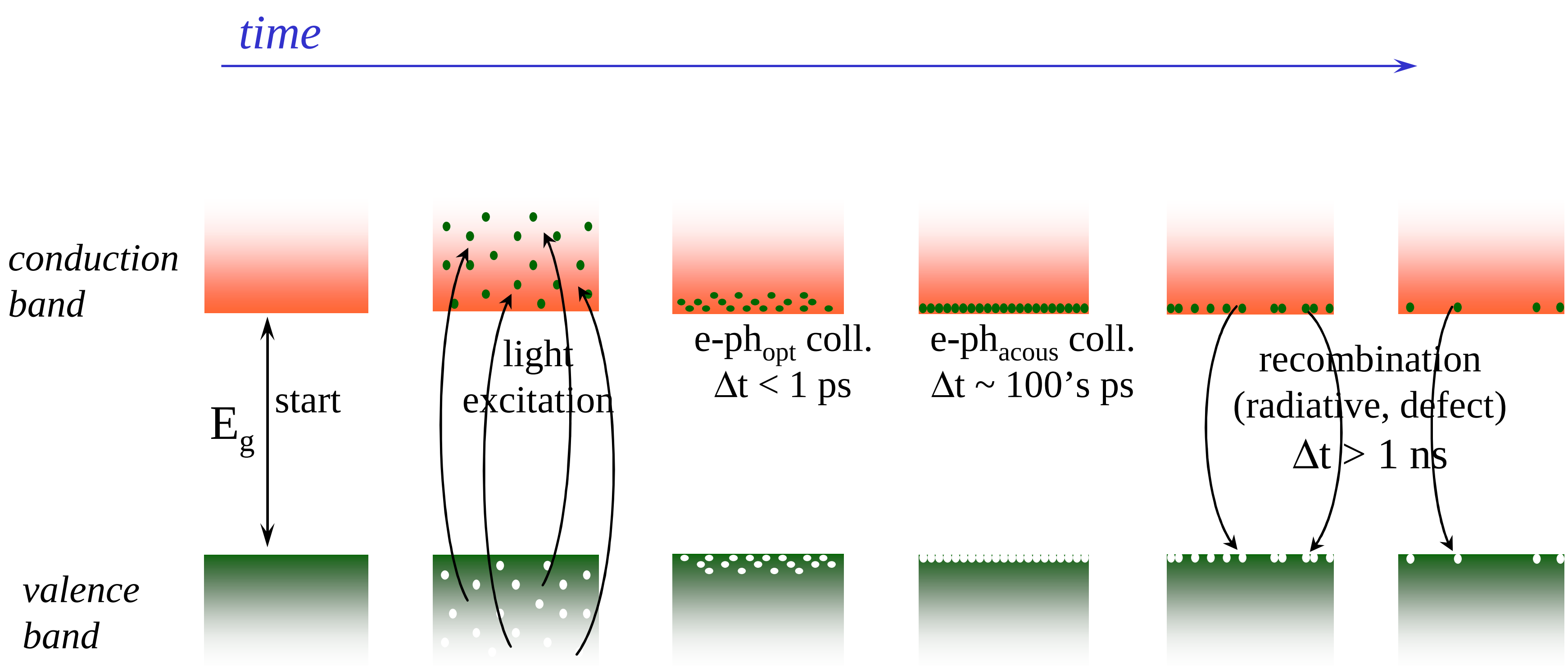
Relaxation time



- Minimal field variation up to $H_{c2}/2$, a factor of two above that
- Mechanism above that unclear. Gap closes with increased field; this will shorten lifetime.

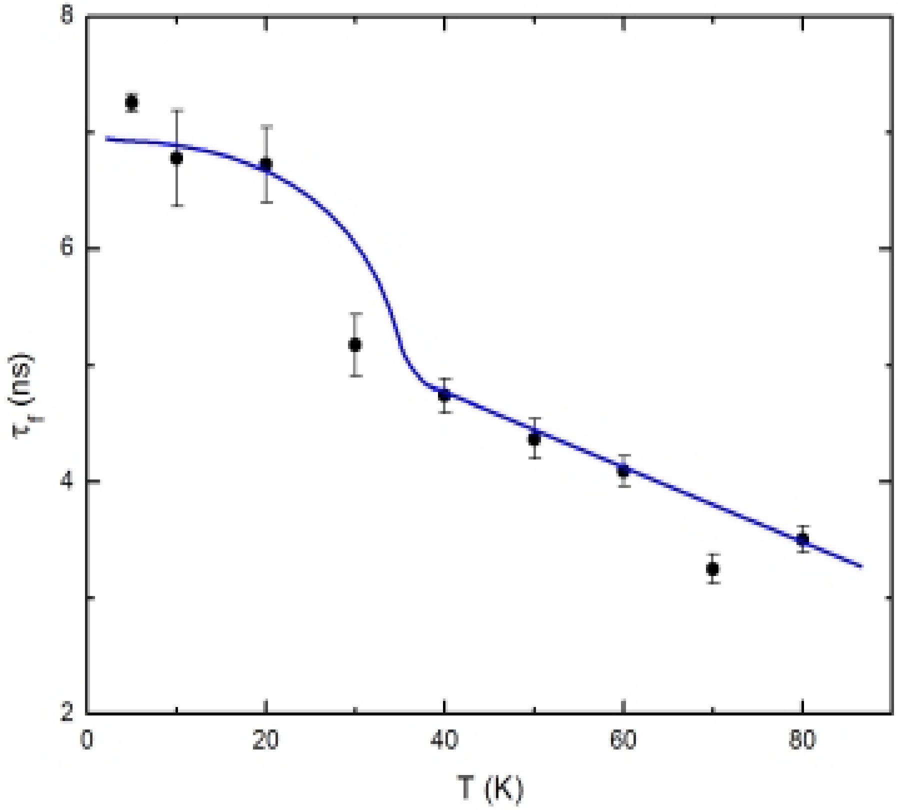
Semiconductor Photoexcitation and Relaxation

Undoped semiconductors at low temperatures



- Far-IR sees the moble free carriers.
- What is the effect of magnetic ordering in DMS?

Decay Parameters

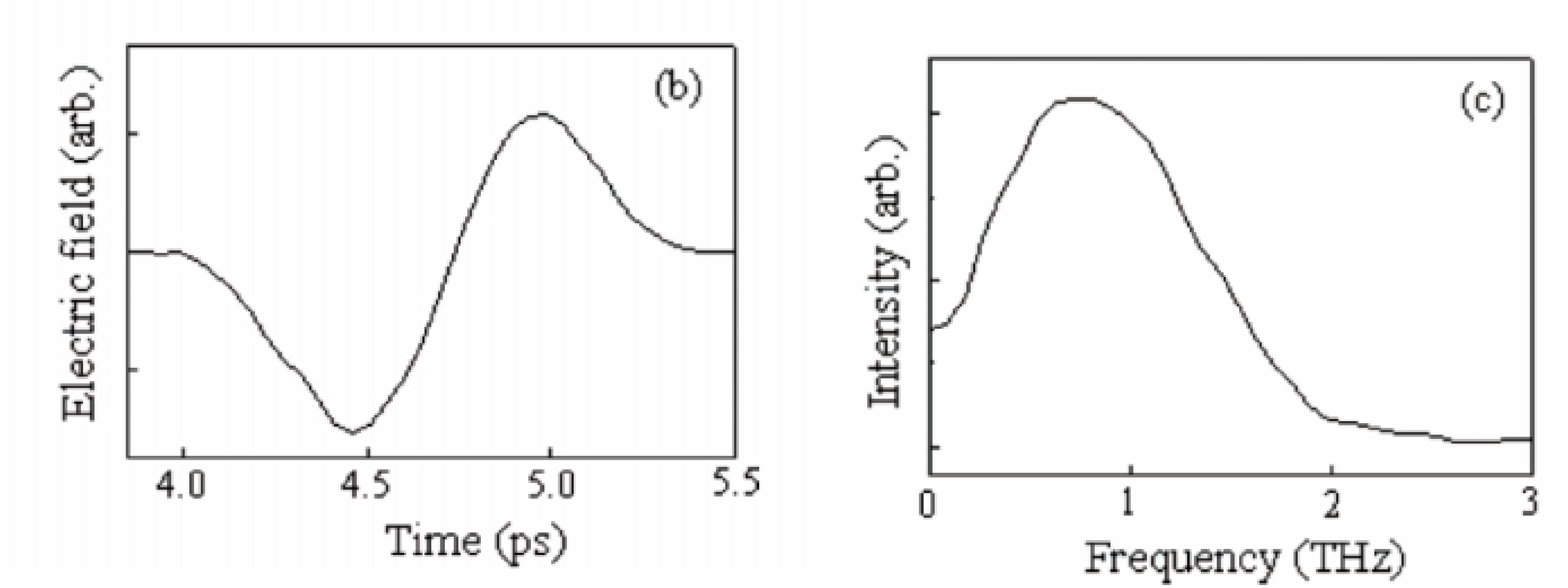


- The decay follows

$$S(t) = \frac{A}{T\sqrt{\pi}} \int_{0}^{+\infty} e^{-\frac{t'}{\tau}} e^{-(\frac{t-t'}{T})^2} dt'$$

- where *S* is the measured temporal response, *A* is the measured signal magnitude, *T* is the probe pulse width, and τ is the effective lifetime.

Very short, very intense pulses



- From SDL (Shen *et al* PRL 2007).
- Equivalent to 1 cycle of THz radiation
- E = 0.7 MV/cm
- Leads to very nonlinear phenomena
  - RF current > j_c in superconductors
  - Hole burning in inhomogeneously broadened transitions
  - Optical rectification in nonlinear materials
  - Domain wall motion in ferroelectrics

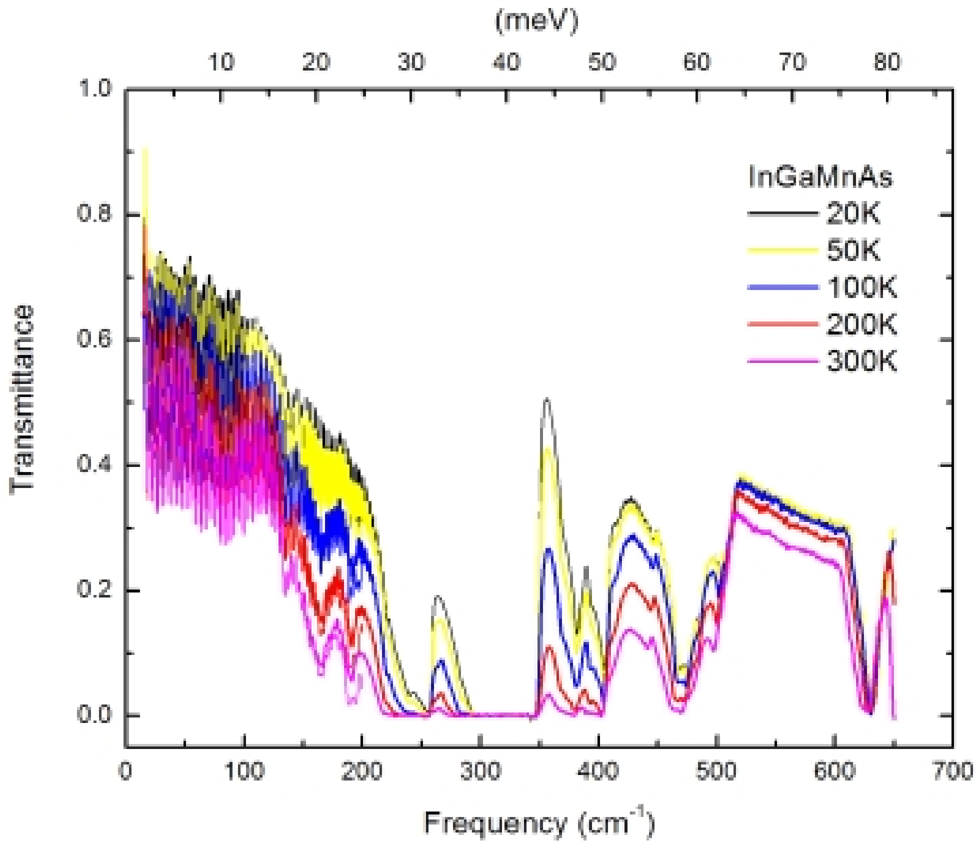
Summary

- Synchrotron is a useful source of FIR radiation
  - –High brightness makes it superior to Hg arc in FIR
- Gives opportunities for very far-IR spectroscopy
- Time structure enables pump/probe studies of many materials.
- Unique feature: broad spectral range of probe: FIR—visible

–Thanks to DOE, grant DE-FG02-02ER45984 at UF and DE-AC02-98CH10886 at the NSLS

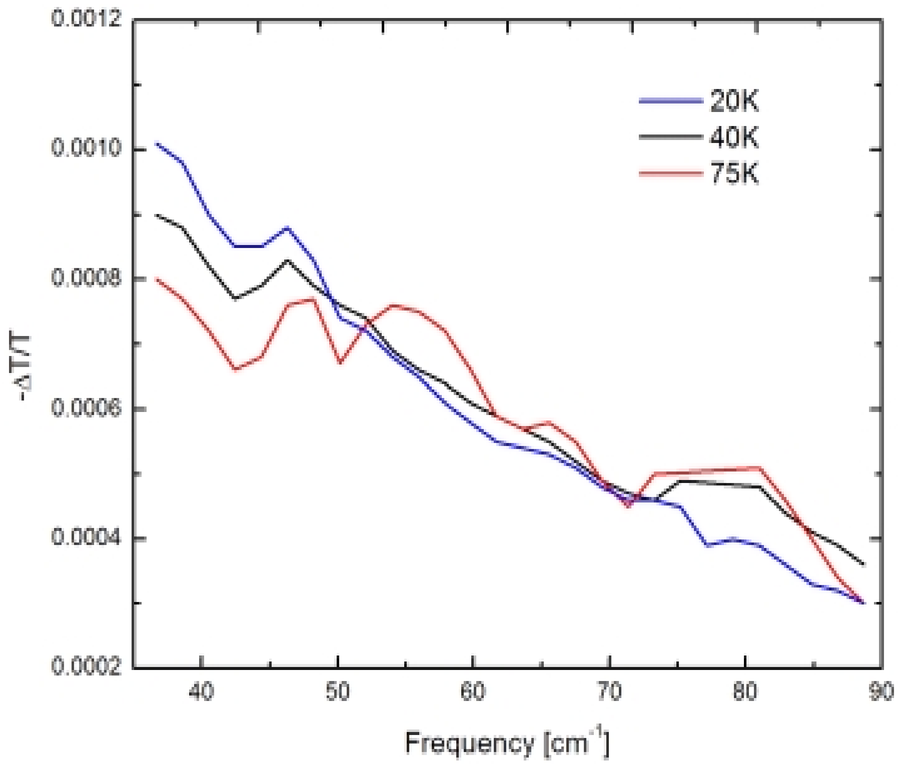
The end

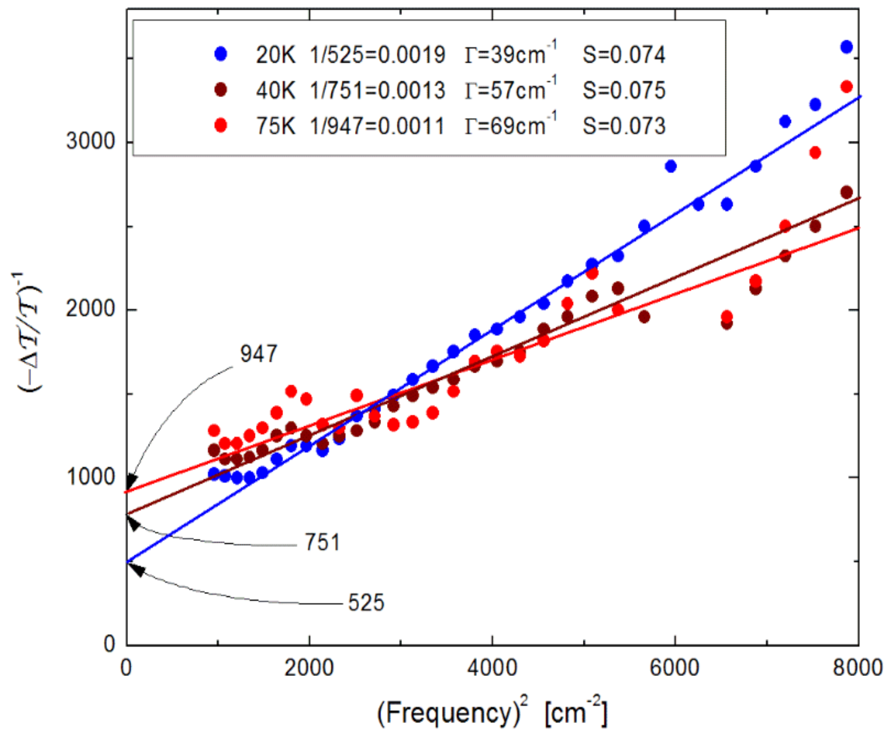
DMS Transmittance



- DMS film: InGaMnAs (50nm) / InGaAs (120nm) / InP(001) (0.4mm)
- ~7.5% Mn doping level
- Ferromagnetically ordered below 40 K

Photoinduced Spectroscopy





- $-\Delta T/T \approx \sigma_1 d = \sigma_{DC} d/(1+\omega^2/\Gamma^2)$, where $S = \sigma_{DC} d \times \Gamma = ne^2 d/m$
- The Drude fit predicts a carrier density $n \sim 7\times10^{15}$ cm^{-3} per laser pulse.
- An estimate of photoinduced carrier density direct from laser pulse is $n \sim 5\times10^{15}$ cm^{-3} .